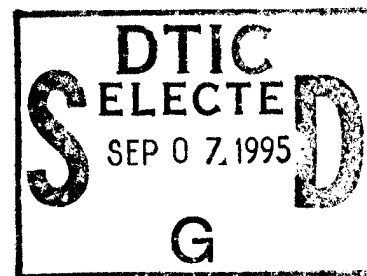




NONLINEAR OPTICAL THIN FILMS

J. R. McNEIL

University of New Mexico
Department of Electrical Engineering
University Hill NE
Albuquerque, NM 87106



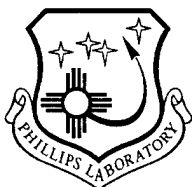
January 1991

Final Report

19950905 013

APPROVED FOR PUBLIC RELEASE; DISTRIBUTION IS UNLIMITED.

DTIC QUALITY INSPECTED 1



PHILLIPS LABORATORY
Lasers and Imaging Directorate
AIR FORCE MATERIEL COMMAND
KIRTLAND AIR FORCE BASE, NM 87117-5776

This final report was prepared by the University of New Mexico, Department of Electrical Engineering, University Hill NE, Albuquerque, NM 87106 under Contract F29601-88-C-0088, Job Order 33261B19 with the Phillips Laboratory, Kirtland Air Force Base, New Mexico. The Laboratory Project Officer-in-Charge was Christopher M. Clayton (LIDN).

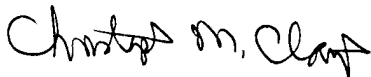
When Government drawings, specifications, or other data are used for any purpose other than in connection with a definitely Government-related procurement, the United States Government incurs no responsibility or any obligation whatsoever. The fact that the Government may have formulated or in any way supplied the said drawings, specifications, or other data, is not to be regarded by implication, or otherwise in any manner construed, as licensing the holder, or any other person or corporation; or as conveying any rights or permission to manufacture, use, or sell any patented invention that may in any way be related thereto.

This report has been authored by a contractor of the United States Government. Accordingly, the United States Government retains a nonexclusive, royalty-free license to publish or reproduce the material contained herein, or allow others to do so, for the United States Government purposes.

This report has been reviewed by the Public Affairs Office and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nationals.

If your address has changed, if you wish to be removed from the mailing list, or if your organization no longer employs the addressee, please notify PL/LIDN, Kirtland AFB, NM 87117-5776, to help maintain a current mailing list.

This technical report has been reviewed and is approved for publication.

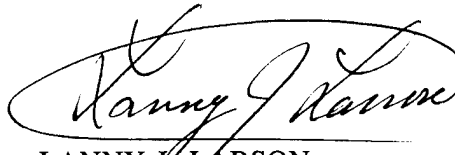


CHRISTOPHER M. CLAYTON
Project Officer



MARC R. HALLADA
Lt Col, USAF
Chief, Laser Systems Division

FOR THE COMMANDER



LANNY J. LARSON
Col, USAF
Director, Lasers and Imaging Directorate

DO NOT RETURN COPIES OF THIS REPORT UNLESS CONTRACTUAL OBLIGATIONS OR NOTICE ON A SPECIFIC DOCUMENT REQUIRES THAT IT BE RETURNED.

REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.				
1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE January 1991		3. REPORT TYPE AND DATES COVERED Final Report: Oct 88 - Dec 90
4. TITLE AND SUBTITLE Nonlinear Optical Thin Films			5. FUNDING NUMBERS C: F29601-88-C-0088 PE: 62601F PR: 3326 TA: 1B WU: 21	
6. AUTHOR(S) J. R. McNeil				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of New Mexico Department of Electrical Engineering University Hill NE Albuquerque, NM 87106			8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Phillips Laboratory Lasers and Imaging Directorate 3550 Aberdeen Ave SE Kirtland Air Force Base, NM 87117-5776			10. SPONSORING/MONITORING AGENCY REPORT NUMBER PL-TR--91-1065	
11. SUPPLEMENTARY NOTES Publication of this report does not constitute approval or disapproval of the ideas or findings. It is published in the interest of STINFO exchange. The established procedures for editing reports were not followed for this technical report				
12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited.			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) The effort consisted of experimental work to deposit and characterize thin films as well as a modeling effort. PZLT materials were emphasized, with several compositions examined. In addition, limited effort was directed toward deposition of other materials, including barium titanate and strontium-barium niobate.				
14. SUBJECT TERMS nonlinear optics, thin films, PLZT, deposition techniques			15. NUMBER OF PAGES 10	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT SAR	

1.0 INTRODUCTION

This is the Final Report for the effort "Nonlinear Optical Thin Films" for the period 1 October, 1988 through 29 December 1990. The effort consisted of experimental work to deposit and characterize thin films as well as a modeling effort. PLZT materials were emphasized, with several compositions examined. In addition, limited effort was directed toward deposition of other materials, including BaTiO₃ and (Sr,Ba)Nb₂O₆.

1.1 EXPERIMENTAL INVESTIGATION

The experimental effort included RF magnetron deposition of PLZT of 7/0/100, 8.6/0/100, 28/0/100, 7.6/70/30 compositions. Additionally, ion beam sputtering was used to deposit 28/0/100, 15/0/100, 7/0/100, and 0/0/100. Many substrate materials were used, including fused silica, sapphire, Si, and Si and GaAs with buffer layers of SiO₂. A major thrust of the ion-beam deposition effort was the growth of films with good crystallinity and orientation on GaAs substrates at temperatures below ~500 °C. This is necessary to prevent degradation of the GaAs material and of any device structures which may be present. This will be important in moving towards integrated PLZT/GaAs photonic devices. This goal was achieved.

We used the following diagnostic techniques to evaluate the films:

X-ray diffraction to determine the crystal structure

Electron dispersion spectroscopy to determine the elemental composition

Alpha-step to determine film thickness

Confocal scanning differential polarization microscopy to determine the film electrooptic properties

Second harmonic generation was obtained using YAG laser output at 1.06 μm as the fundamental wavelength. We investigated the angular dependence of the intensity of the SHG from the films to determine components of the nonlinear optical tensor.

Waveguiding was achieved in PLZT in structures of ITO/PLZT/SiO₂/Si and ITO/PLZT/SiO₂/GaAs. By applying an electric field of 0 to 300 V/mm to the ITO and Si substrate, and using prism out-coupling, we observed modal angular shifts. The total angular shift was due primarily to the contribution from a positive change in the effective index of the film which was likely the result of changing the thickness of the film. The contribution from pure electric-field-induced birefringence change is small and reduces the index. Both TE and TM modes exhibit similar shifting behaviour.

We investigated the time evolution of the carrier distribution and induced electric field in a photorefractive material in the presence of a cw laser field and a pulsed electric field. Some effort was spent investigating the effects of the photovoltaic term in both the steady state and the time dependent situations. In addition, we investigated the high-speed switching characteristics of PLZT and observed the material to a response time of less than 2 nsec.

Plasma etching was examined using chlorinated and fluorinated plasmas produced using instrumentation-limited dc hollow cathode discharges. Etch rates of PLT thin

<input checked="checked" type="checkbox"/>	
<input type="checkbox"/>	
<input type="checkbox"/>	
Codes	
Dist	Avail and/or Special
A-1	

films as high as 650 nm/hour were obtained. After etching, the stoichiometry was characterized to indicate that in some situations (e.g. elevated substrate temperature) significant changes occur. This is significant if the material is to be applied to construct electronic and optoelectronic devices.

1.2 THEORETICAL INVESTIGATION

The theoretical efforts were a relatively small portion of this project. The modeling was carried out by Chibing Xu, under the supervision of Professor McIver and constituted a major portion of his PhD dissertation. This work has also resulted in two completed manuscripts that have been submitted for publication in the Journal of the Optical Society of America B.

The subject of these manuscripts is the response of a photorefractive material to a cw gaussian laser beam and a transverse electric field. This work was motivated by the frequency doubling experiments performed in the Center for High Technology Materials on thin film PLZT materials. We were particularly interested in the response of the photogenerated carriers to a time-dependent externally applied electric field.

The first paper explores the steady state behavior of a photorefractive material subjected to a cw gaussian laser beam. The photorefractive material is described by the phenomenological band transport model of Kukhtarev. In order to simplify the mathematics the calculations were carried out in one dimension and the photovoltaic term was ignored. The latter approximation is both common and justified for most materials. Unlike other calculations, we did not linearize the equations. As can be seen in the attached manuscript the nonlinear terms can be important at relatively low intensities for some materials.

The second paper extends the results of the first paper by adding a transverse electric field to the material. In some cases this field is periodically turned on and off thereby simulating the conditions under which the frequency doubling experiments were performed. These calculations showed that the external electric field in the illuminated region of the material was reduced because of the external field, that the relaxation time of the electric field within the materials depends strongly on the intensity of the light, and that the presence of an oscillating electric field can enhance the induced electric field.

2.0 PUBLICATIONS

The following publications resulted from this effort:

1. C. Xu, D. Statman, and J. K. McIver, "Effects of a Gaussian Beam on Photorefractive Materials: I. Steady State Solution", Submitted to Journal of Optical Society of America B.
2. C. Xu, J. K. McIver, and D. Statman, "Effects of a Gaussian Beam on Photorefractive Materials: II. Dynamic Behavior Under Externally Applied Electric Field", Submitted to Journal of Optical Society of America B.
3. F. Wang, C.-B. Juang, C. Bustamante, and A. Y. Wu, "Electro-Optic Properties of (Pb,Lu)(Zr,Ti)O₃, BaTiO₃, (Sr,Ba)Nb₂O₆ and Ba₂NaNb₅O₁₅ Thin Films by a Confocal

Scanning Differential Polarization Microscope", in the 4th International SAMPE Electronics Conference, Volume 4, Electronic Materials-Our Future, pp. 712-721, ed. R. E. Allred, R. J. Martinez, and K. B. Wischmann, (Society for the Advancement of Materials and Process Engineering) 1990.

4. A. Y. Wu, "Deposition of (Pb,La)(Zr,Ti)O₃, BaTiO₃, (Sr,Ba)Nb₂O₆, Ba₂NaNb₅O₁₅, KTiOPO₄, and beta-BaB₂O₄ Thin Films", in the 4th International SAMPE Electronics Conference, Volume 4, Electronic Materials-Our Future, pp. 722-733, ed. R. E. Allred, R. J. Martinez, and K. B. Wischmann, (Society for the advancement of Materials and Processing Engineering) 1990.

5. M. R. Poor, A. M. Hurd, C. B. Fleddermann, and A. Y. Wu, "PLZT Thin Film Etching using Plasma Techniques", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

6. A. Y. Wu, F. Wang, C.-B. Juang, and C. Bustamante, "2-D High Definition and High Resolution PLZT Thin Film Spatial Light Modulators", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

7. A. Y. Wu, F. Wang, C.-B. Juang, C. Bustamante, C.-Y. Yeh, and J.-C. Diels, "Electro-Optic and Non-Linear Optical Properties of (Pb,La)(Zr,Ti)O₃, BaTiO₃, (Sr,Ba)Nb₂O₆, Ba₂NaNb₅O₁₅, and beta-BaB₂O₄", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

8. F. Wang and A. Y. Wu, "Electro-Optical and Nonlinear Optical Properties of Thin Film Materials Containing Oxygen-Octahedra Under High DC Electric Field", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

9. A. Mukherjee, S. R. J. Brueck, and A. Y. Wu, "Electric Field Induced Second Harmonic Generation in PLZT", Opt. Commun. 76(3,4) 220 (1990).

10. A. Y. Wu, F. Wang, C.-B. Juang, and C. Bustamante, "Electro-Optic and Non-Linear Optical Coefficients of (Pb,La)(Zr,Ti)O₃, BaTiO₃, (Sr,Ba)Nb₂O₆, and Ba₂NaNb₅O₁₅ Thin Films", in the Symposium on Ferroelectric Thin Films, Spring Meeting, Materials Research Society, San Francisco, California (1990). (In press)

11. M. R. Poor, A. M. Hurd, C. B. Fleddermann, and W. Y. Wu, "Plasma Etching of PLZT and PLT using Chlorine Based Gases", in the Symposium on Ferroelectric Thin Films, Spring Meeting, Materials Research Society, San Francisco, California (1990). (In press)

12. L.L. Boyer, A. Y. Wu, and J. R. McNeil, "Physical and Electro-Optical Properties of Ion Beam Sputtered Thin Film PLZT Ceramic", in the Symposium on Ferroelectric Thin Film, Spring Meeting, Materials Research Society, San Francisco, California (1990). (In press)

13. A. B. Wegner, S. R. J. Brueck, and A. Y. Wu, "Integrated PLZT Thin Film Waveguide Modulators", Ferroelectrics, 116, 195 (1991).

14. A. Mukherjee, S. R. J. Brueck, and A. Y. Wu, "Electro-Optic Effects in Thin Film Lanthanum-doped Lead Zirconate Titanate", Opt. Lett. 15(3) 151 (1990).

15. L. L. Boyer, A. Y. Wu, G. W. Metzger, and J. R. McNeil, "Properties of Ion Beam Deposited PLZT", J. Vac. Sci. Tech. A7 (3), 1199 (1989).
16. Feiling Wang and A. Y. Wu, "An Analytical Model for the Quadratic Electrooptic Effect of Perovskites", Submitted to Physical Review Letters.

PL-TR--91-1065

DISTRIBUTION

AFSAA/SAI, Washington DC
AUL/LSE, Maxwell AFB, AL
DTIC/OCC, Alexandria, VA
PL/SUL/HO, Kirtland AFB, NM
Official Record Copy, (PL/LIDN, Dr Chris Clayton)